

O 22: Theoretical methods

Time: Monday 17:15–19:30

Location: WIL C107

O 22.1 Mon 17:15 WIL C107

High Temperature Thermal Conductivity from First Principles — ●CHRISTIAN CARBOGNO^{1,2}, RAMAMURTHY RAMPRASAD^{1,3}, and MATTHIAS SCHEFFLER¹ — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4–6, 14195 Berlin, Germany — ²Materials Department, University of California Santa Barbara, USA — ³Chemical, Materials & Biomolecular Engineering, University of Connecticut, Storrs, USA

In spite of significant research efforts, little is yet known about the atomistic details and mechanisms that underlie peculiarly low (or high) thermal conductivities, especially at elevated pressures and temperatures. Under such extreme conditions, systematic experimental measurements are hard to perform; conventional theoretical approaches typically fail to capture significant physical aspects of the problem, since these methods are either inherently limited to (a) low temperatures and/or (b) to perfect crystals. A recently developed *ab initio* simulation strategy [1] allows to overcome the latter limitation, but the assessment of the high temperature regime remains an unsolved challenge. Within this work, we present efficient strategies to overcome this serious restriction and show their applicability for zirconia based ceramics – a material typically used in high temperature applications, for instance in thermal barrier coatings [2].

[1] T. M. Gibbons, and S. K. Estreicher, *Phys. Rev. Lett.* **102**, 255502 (2009).

[2] D. R. Clarke, and C. G. Levi, *Annu. Rev. Mat. Res.* **33**, 383 (2003).

O 22.2 Mon 17:30 WIL C107

Representing Potential-Energy Landscapes by High-Dimensional Neural Networks — NONGNUCH ARTRITH, TOBIAS MORAWIETZ, and ●JÖRG BEHLER — Lehrstuhl für Theoretische Chemie, Ruhr-Universität Bochum, 44780 Bochum, Germany

The construction of efficient interatomic potentials is a necessary condition for carrying out large scale molecular dynamics and Monte Carlo simulations. However, the identification of a suitable functional form to represent a given set of reference data from *ab initio* calculations with high accuracy is a formidable challenge.

Using several benchmark systems, we show that flexible artificial neural networks (NNs) are able to provide reliable high-dimensional potential-energy surfaces even if complex bonding situations are present. They can be applied to a wide range of systems from bulk metals and surfaces to small molecules. The scope and limitations of NN potentials are discussed, and the accuracy is compared with electronic structure calculations.

O 22.3 Mon 17:45 WIL C107

Stress formulation within the FLAPW method

— ●AARON KLÜPPELBERG, GUSTAV BIHLMAYER, and STEFAN BLÜGEL — Peter Grünberg Institut & Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

Stress is an important quantity in characterizing the states of condensed matter. It can be modified by epitaxial growth relation or application of electric or magnetic fields. This makes the computation of the stress field particularly important for understanding the properties and functionality of ferroic and multiferroic oxides.

The full-potential linearized augmented plane wave (FLAPW) method is an all-electron electronic structure method based on the density functional theory (DFT) that is well suited to describe electronically and structurally complex materials. It is realized in the FLEUR code [1], where orbital-dependent functionals [2] are implemented to describe the exchange-correlation energy of oxides.

We present here a stress-strain formalism for elastic deformations using the frozen augmentation approximation. It takes into account the Pulay corrections to the Hellmann-Feynman stress that appear due to the use of an incomplete basis set (IBS). This procedure allows a simultaneous relaxation of lattice shape and atom positions. Furthermore, this formalism enables the application of pressure and shear making an investigation of phase transitions due to external mechanical impact accessible.

[1] www.flapw.de

[2] M. Betzinger, C. Friedrich, and S. Blügel, *PRB* **81**, 195117 (2010).

O 22.4 Mon 18:00 WIL C107

Massively Parallel Real-Space DFT Calculations — ●PAUL BAUMEISTER¹, DANIEL WORTMANN¹, TOMOYA ONO², and STEFAN BLÜGEL¹ — ¹Peter Grünberg Institut & Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany — ²Dept. of Precision Engineering, Osaka University, Osaka, Japan

We present a new DFT tool developed in Jülich and Osaka that combines equidistant real-space grids and the Projector Augmented Wave (PAW) method. The code is explicitly designed for the structural relaxation of systems consisting of several thousands of atoms with very flexible boundary conditions. The specific advantage of our approach lies in the possibility to achieve a high degree of parallelization. The real-space treatment of wave functions, densities and potentials enables a simple and efficient parallelization with respect to communication and load balancing. Besides the parallel computation of k-points, we employ a domain decomposition to the real-space cell and a parallelization of eigenvalues (bands). Hence, we exploit the local character of the Kohn-Sham equation and approximate the kinetic energy operator by finite differences, avoiding Fourier transforms completely. The arising communication pattern suits perfectly the data network of IBMs BlueGene type massively parallel supercomputers. We will discuss the challenges of band parallelization, in which communication cannot be avoided as much as in k-points or even the real-space domain decomposition. Besides the demonstration of the efficiency of the parallelization, we will also report on the obtainable accuracy.

O 22.5 Mon 18:15 WIL C107

Tight-Binding Parameters from DFT Calculations: First Applications to Hydrocarbons — ●ALEXANDER URBAN¹, MARTIN REESE^{2,3}, MATOUS MROVEC^{2,3}, CHRISTIAN ELSÄSSER^{2,3}, and BERND MEYER¹ — ¹ICMM/CCC, University of Erlangen-Nürnberg, Germany — ²Fraunhofer IWM, Freiburg, Germany — ³IZBS, Karlsruhe Institute of Technology, Germany

Tight-binding (TB) models for practical calculations are usually derived by fitting band structures and total energies to results of DFT calculations or experimental data. We have developed an alternative approach in which the TB parameters are determined directly from DFT data for arbitrary reference configurations without extensive fitting. Our method [1] is conceptually different from previous approaches [2] as it is based on a projection [3] of basis-set-converged wave functions from mixed-basis DFT computations onto a minimal basis of atomic orbitals. The radial shape of the atomic orbitals is optimized by minimizing the loss (spillage) in the projection procedure. The Slater-Koster tables are then calculated with the optimized minimal basis using the self-consistent DFT Hamiltonian. To demonstrate the quality of the derived TB models results for the electronic structures, total energies and forces for different carbon and hydrocarbon systems will be compared to DFT reference data.

[1] A. Urban, M. Reese, M. Mrovec, C. Elsässer, B. Meyer, submitted

[2] D. Porezag et al., *Phys. Rev. B* **51** (1995) 12947

[3] D. Sanchez-Portal et al., *Sol. State Comm.* **95** (1995) 685

O 22.6 Mon 18:30 WIL C107

Localized resolution of identity for efficient Hartree-Fock exchange, based on numeric atom-centered orbitals — ●JÜRGEN WIEFERINK, VOLKER BLUM, XINGUO REN, PATRICK RINKE, and MATTHIAS SCHEFFLER — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Methods based on an exact exchange operator (EX) are increasingly popular, but are still restricted to analytical basis functions (e.g. Gaussians) for medium system sizes. We here introduce a localized resolution-of-identity approach for the two-electron Coulomb operator, based on expanding single-particle basis function products separately into auxiliary atom-centered basis sets that are restricted to two centers. Our approach produces accurate results for all-electron EX, can be applied both to analytical and numeric basis functions, requires only $\mathcal{O}(N^2)$ intermediate storage and retains a path towards $\mathcal{O}(N)$ EX for large systems. We demonstrate a total-energy accuracy of < 1 meV/atom for systems including Alanine chains and the S22 benchmark molecule set [1], using the numeric atom-centered orbital based all-electron electronic structure code FHI-aims [2].

[1] P. Jurečka et al., *Phys. Chem. Chem. Phys.* **8**, 1985 (2006).

[2] V. Blum *et al.*, Comput. Phys. Comm. **180**, 2175 (2009).

O 22.7 Mon 18:45 WIL C107

Accurate Calculation of the Single-Site Green Function in Relativistic Full-Potential Scattering — ●PASCAL KORDT, RUDOLF ZELLER, PHIVOS MAVROPOULOS, and STEFAN BLÜGEL — Peter Grünberg Institut & Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

Within the Korringa-Kohn-Rostoker (KKR) method the Green function for a single scattering centre can be calculated using the regular and irregular solutions of the corresponding angular Lippmann-Schwinger equation. We have developed a method for a relativistic treatment valid also for non-spherical potentials. It is based on an expansion in Chebyshev polynomials and their recursion relations. This allows us to rewrite the Lippmann-Schwinger integral equation into a system of algebraic linear equations. The computational effort for solving these equations is drastically reduced by dividing the radial integration into sub-intervals with a suitable matching technique. Compared to previous perturbation methods a much higher accuracy is achieved with modest increase in computational effort.

O 22.8 Mon 19:00 WIL C107

Wavefunction-based correlation method for metals: from bulk to surfaces and interfaces — ●ELENA VOLOSHINA and BEATE PAULUS — Institut für Chemie und Biochemie, Freie Universität Berlin, 14195 Berlin, Germany

Theoretical determination of surface energies is a great challenge. While density functional theory (DFT) provides understanding of qualitative trends, absolute values depend strongly on the choice of the functional. An alternative approach with the advantage of the possibility of a systematical improvement are the wavefunction-based methods. One problem here is that a very accurate treatment, e.g. with the coupled-cluster method, scales very unfavorably with the number of electrons in the system. For this reason the method of increments was invented to calculate the correlation energy of bulk metals in terms of contributions from localized orbital groups [1,2]. This approach can be

extended to system of low dimensionality, like surfaces. Furthermore, the method of increments is useful when considering surface adsorption phenomena, since DFT often fails to describe physisorption correctly. To demonstrate our approach we selected Mg(0001) surface, where the method of increments is used to calculate surface energy as well as Xe adatom interaction energy.

[1] E. Voloshina, N. Gaston, and B. Paulus, J. Chem. Phys. **126**, 13411(2007); [2] E. Voloshina and B. Paulus: Phys. Rev. B **75**, 245117 (2007); Chem. Modell. **6**, 162 (2009); Z. Phys. Chem. **224**, 369 (2010).

O 22.9 Mon 19:15 WIL C107

Exchange an Correlation effects in photoemission spectroscopy: from semiconductors to transition metal oxides — ●MATTEO GUZZO¹, MATTEO GATTI², FRANCESCO SOTTILE¹, PINA ROMANIELLO¹, FAUSTO SIROTTI³, JOHN REHR⁴, and LUCIA REINING¹ — ¹LSI - ETSF, Ecole Polytechnique, Palaiseau, France — ²ETSF, Universidad del Pais Vasco, San Sebastian, Spain — ³Synchrotron Soleil, Gif-sur-Yvette, France — ⁴University of Washington, Seattle WA, USA

As a prototype of strongly-correlated systems, NiO has been extensively studied, however the origin of its gap is still under debate. Many-Body Perturbation Theory (MBPT) (with the one-particle Green's function, *aka* the propagator of a particle, as key quantity) describes appropriately *ab-initio* photoemission gaps and spectra. *GW* approximation from MBPT includes dynamical correlation effects beyond H-F. The standard *GW* approach is not reliable on NiO since the Kohn-Sham band structure is too poor a starting point. COHSEX — a static approximation of *GW* — allows one to achieve full self-consistency, and to obtain a better starting point for the *GW* step. The COHSEX+*GW* result gives about 5 eV energy-gap, in good agreement with experiment and with other approaches beyond G_0W_0 . An attempt to apply *vertex corrections* from TDDFT has shown no improvement in the spectrum. Here we focus on the effects of vertex corrections on quasiparticle energies and satellites and compare the cases of Si and NiO. The Cumulant Expansion approach is discussed as a possible alternative approach.